



Laser Probe Makes First Gas Species Measurements in Pool Fire

As the culmination of a three-year Laboratory Directed Research and Development (LDRD) project, gas species and soot were measured simultaneously in 1-meter-diameter pool fires fueled by JP-8, the jet fuel used by the U.S. Air Force. These proof-of-principle experiments were conducted at Sandia-New Mexico's Burn Site, in an enclosed facility known as the Fire Laboratory for Accreditation of Models and Experiments (FLAME, see Figure 1). The CRF's Chris Shaddix and Doug Scott conducted the pool fire experiments, together with technologists at the Burn Site. The development of the tunable diode laser (TDL) diagnostic and the associated fiber-coupled, water-jacketed optical probe was led by Chris and involved the talents of a number of Sandians, especially Sarah Allendorf, Dave Ottesen, Howard Johnsen, Jimmy Ross, Gary Hubbard (of Hubbard Associates), Sal Birtola, Phil Santangelo (now at Micron Optics), and Peter Ludowise (now at 3M). Lou Gritz, manager of Sandia's Fire Science and Technology Department, also contributed to the project.

Researchers at Sandia have been investigating large-scale pool fires, both experimentally and numerically, for several years, because of the risk that these fires could pose to critical engineered systems in transport accidents. Fielding laser or optical diagnostics in this turbulent, optically thick, high-temperature environment is difficult. Therefore, data capture in large fires has traditionally been limited to relatively crude, poorly resolved techniques, such as those that use



Figure 1. The Sandia laser probe is shown at the beginning of a 1-m-diameter, JP-8 pool fire conducted at Sandia's Lurance Canyon Burn Site in Albuquerque, New Mexico. The horseshoe-shaped probe head, which contains the optical components, is side-supported and hangs down into the fire so that flame measurements can be made in the space between the light pipes attached to the ends of the horseshoe.

thermocouples and heat-flux gauges. However, in situ laser measurements with high spatial and temporal resolution are needed to help guide the development of simplified models of combustion and soot formation for incorporation into supercomputer simulations of large fires.

As part of this project, advances were made in high-frequency multiplexing of the diode lasers. Modulation frequencies of ~ 1 MHz are being used, in concert with second harmonic (2f) detection with custom-built lock-in amplifiers with an 8- μ s time constant. This setup allows spectral sweeps of the lasers at a 1-kHz repetition rate. In addition, an open-path Herriott cell optical design was incorporated into

the probe head, allowing the near-infrared TDL light to traverse the probe volume 18 times before being captured onto a fiberoptic return cable. This optical multipassing is necessary in order to have sufficient laser absorption sensitivity for the weak overtone molecular absorption transitions accessible in the near-IR. Holes drilled through the centers of the Herriott mirrors allow red laser extinction measurements (for soot concentration) and near-IR emission measurements (for soot temperature).

Jeff Murphy analyzed the datasets, which entailed fitting the second derivative of a Lorentzian profile to the measured 2f signal for each individual laser sweep. For the initial test series at

FLAME, commercially available distributed feedback TDLs at 1.31 and 1.54 μm were used to detect water vapor and acetylene, respectively. Figure 2 shows a sample time record of soot concentrations and relative water and acetylene concentrations within a pool fire. The dominant “puffing” frequency of ~ 1 Hz is evident in the periodicity of the peaks.

Follow-on funding of this work is allowing the construction of a set of circuit boards that will simultaneously control and detect the signals from 4 multiplexed TDLs, allowing CO and methane TDLs to be added to the instrument. An effort is also being made to improve the quantification of TDL-measured gas concentrations. Four test series in 2-m diameter JP-8 fires are planned for the coming year.

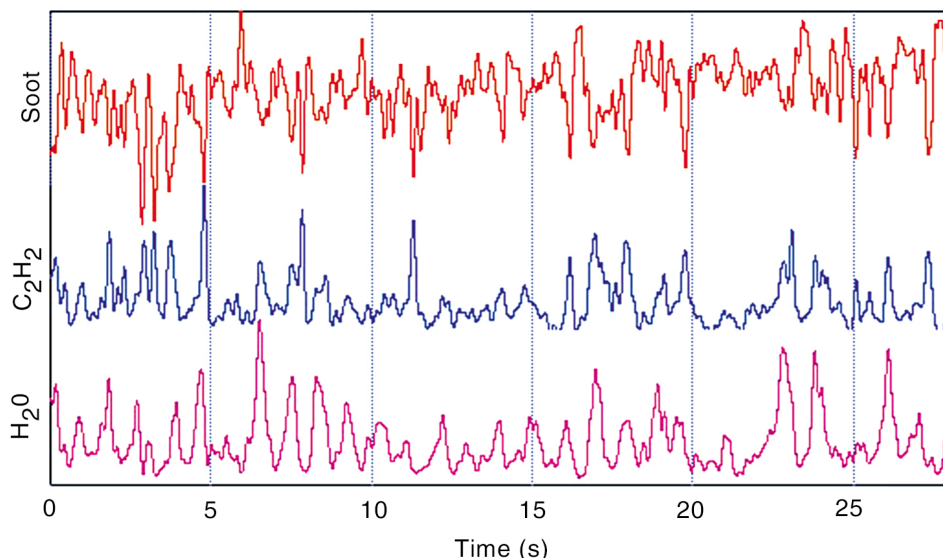


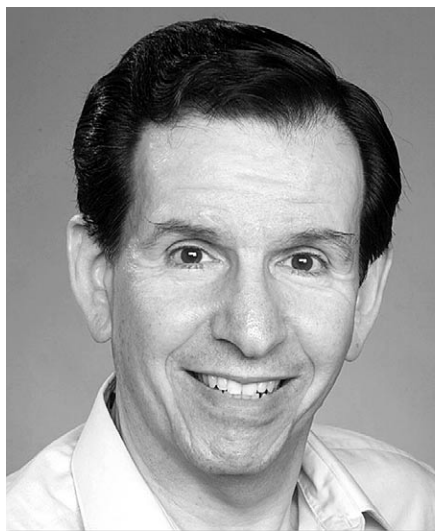
Figure 2. Soot absorption and TDL measurements of water and acetylene for one of the datasets. One can see some correlation among the measurements, notably the inverse correlation between soot and acetylene early in the time series (2–5 s) and the positive correlation between acetylene and water later in the time series (22–25 s).

Awards

Kerstein Elected APS Fellow

In their September 2001 meeting, the Executive Board of the American Physical Society elected Alan Kerstein a Fellow. In electing Alan a Fellow, the Board cited his original contributions to physics as well as his technical leadership.

During his career at the CRF, Alan has modeled, among other things, char oxidation, desorption of gases from coal, percolation in burning propellants, and the action of ocean waves. Most recently, he has been applying one-dimensional turbulence models to a host of research topics. Current projects include fundamental studies of flames, engine simulation, variable-density flow dynamics models, and near-wall modeling for large-eddy simulations. He also collaborates with groups studying incineration (University of Arizona), burning in supernovae (UC Santa Cruz), and soot dynamics in flames (Purdue University).



Alan Kerstein has been elected as a Fellow of the American Physical Society (APS). The Executive Board cited Alan's "substantial and enduring original contributions to turbulence dynamics, turbulent mixing, and turbulent combustion" as well as his "insightful technical leadership among peers and students." Election is a singular honor, bestowed on no more than one half of one percent of APS members.

Engine Group Members Capture SAE Awards

Dennis Siebers and John Dec won awards for Excellence in Oral Presentation at the SAE 2001 World Congress, held March 5–8 in Detroit, Michigan. Dennis presented a paper entitled "Flame Lift-Off on Direct-Injection Diesel Sprays Under Quiescent Conditions" (summarized in the August/September issue of the CRF News). John's presentation described work done with Dale Tree of Brigham Young University in obtaining OH (PLIF) and soot (PLII) images of the fuel jet impinging on the piston bowl wall. The companion paper, presented by Dale Tree, which also won an award of excellence, described measurements of the soot/wall deposition and its potential as a contributing pathway to engine-out particulate emissions.

Detection of the Vinyl Radical and Acetylene by Laser-Induced Fragmentation Fluorescence

The vinyl radical (C_2H_3) is believed to be an important combustion intermediate in rich hydrocarbon flames. In aliphatic fuel combustion, reactions of vinyl radicals with unsaturated hydrocarbons may contribute to the formation of the first aromatic rings, polycyclic aromatic hydrocarbons, and eventually soot. Despite its importance, the vinyl radical has not been detected in flames by optical methods such as planar laser-induced fluorescence (PLIF) because it has no known fluorescent excited state. In fact, there are presently no demonstrated PLIF schemes for probing any of the species C_2H_n ($n = 2-6$) in the reaction sequence connecting ethane to acetylene.

David Osborn and Jonathan Frank have investigated laser-induced fragmentation fluorescence (LIFF) as a potential diagnostic technique for measuring the vinyl radical and acetylene. A pulsed UV laser was used to photodissociate acetylene and vinyl radicals to C_2 , and the resulting C_2 fluorescence emission was studied. This investigation was motivated by an observation of Jason Rehm and Phil Paul that a 230-nm laser beam produces laser-generated C_2 fluorescence in non-sooting laminar premixed methane/air flames. The fluorescence signal arises from the well known Swan bands of the excited C_2 system. Figure 1 shows the energy level diagram for LIFF processes for acetylene and vinyl radical.

As an initial step towards understanding this process, experiments were carried out in

the more easily controlled environment of a low-pressure photodissociation cell through which reagents continuously flow. Acetylene (C_2H_2) and vinyl radicals (C_2H_3) were found to be the species most likely to fragment to fluorescent C_2 molecules from a group of seven candidates (CH_4 , C_2H_2 , C_2H_3 , C_2H_4 , C_2H_6 , C_3H_6 , and C_4H_6).

Fluorescence spectra from acetylene and vinyl radical (generated from the photolysis of methyl vinyl ketone) are shown in Figure 2. The signal centered at 468 nm is easily assigned to the Swan system, while the CO is produced as a product of methyl vinyl ketone photolysis. This simultaneously collected CO LIF signal provides a useful calibration of the vinyl radical number density. Because the vinyl radical is generated via photolysis, it is born with large amounts of vibrational energy, cooling as it collides with N_2 in the cell. For identical number densities, the vinyl radical was found to be more efficient than acetylene at producing C_2 fluorescence by a factor ranging from 1300 to 200, decreasing in efficiency with decreasing internal excitation of the vinyl radical.

The mechanism suggested to explain the results is shown in Figure 1. Each molecule loses successive H atoms on the way to forming excited C_2 . Considering that the vinyl mechanism passes through a highly excited C_2H_2 intermediate, it might seem surprising that the vinyl radical has a higher LIFF efficiency than acetylene. The key difference between the two

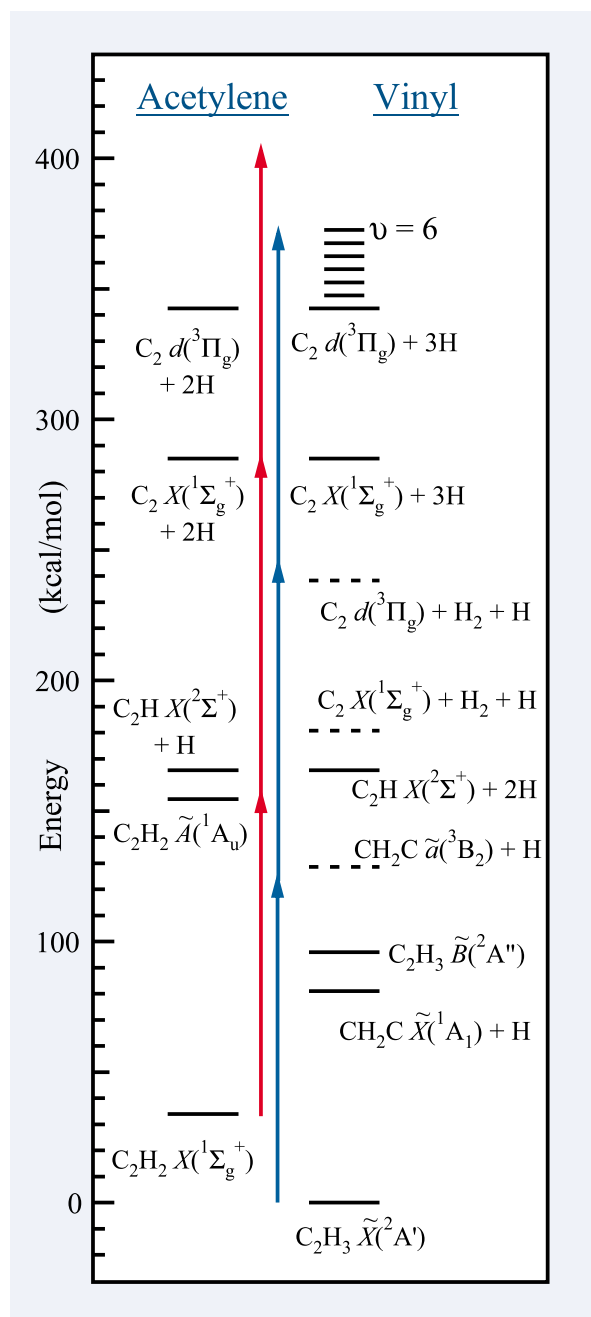
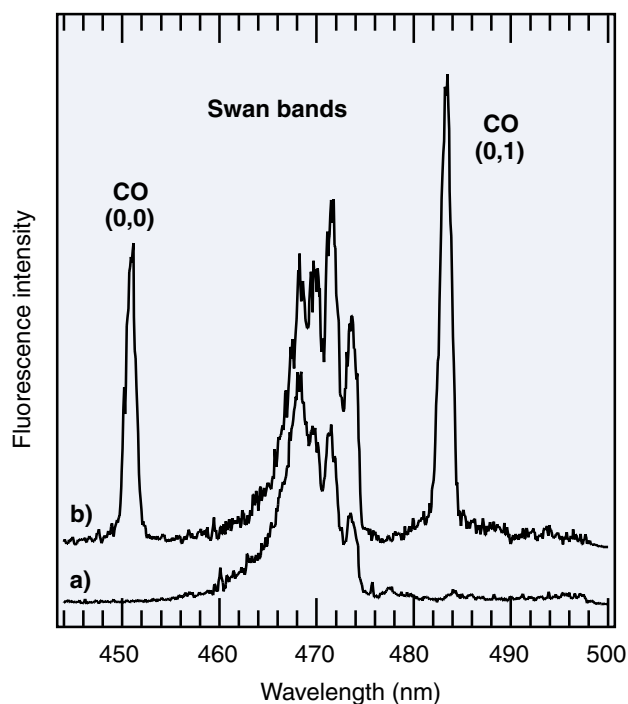


Figure 1. The energy level diagram for 230-nm multiphoton dissociation of acetylene (C_2H_2) and vinyl radical (C_2H_3) shows the steps leading to C_2 formation. For acetylene dissociation, concerted elimination to form $C_2 d(^3\Pi_g) + H_2$ is thermodynamically feasible with two photons, but spin forbidden. The lowest spin-allowed pathway for $C_2 d(^3\Pi_g)$ production requires three photons at 230 nm ($E_{hv} = 124$ kcal/mol).

dissociation mechanisms lies in the loss of the first H atom. For vinyl, a strong, one-photon absorption leads to H atom loss, while for acetylene the first photon absorption is at least three orders of magnitude weaker.

In addition, the C—H bond in acetylene is so strong that two photons are required for bond fission, compared to the vinyl radical C—H bond, which is four times weaker, and can be broken with a single photon. The results show that LIFF may provide an indirect but selective optical diagnostic for the vinyl radical in methane flames and for non-fluorescent species in general.

Figure 2. Fluorescence spectra probed at $\lambda = 230.11$ nm for a) C_2H_2 at 1 mJ/pulse and b) the C_2H_3 and CO products of methyl vinyl ketone photolysis at 0.4 mJ/pulse and 0.5 μ s pump-probe delay. The peaks centered at 468 nm in both spectra are C_2 Swan bands ($d^3\Pi_g \rightarrow a^3\Pi_u$). Trace b) is vertically offset for clarity.



Dave Ottesen (l) and Sarah Allendorf (r) get together to wish Post Doctoral researcher Ben Chorpeneh well in his new job as a staff member at the National Energy Technical Laboratory in Morgantown, West Virginia. While at the CRF, Ben helped develop mid-infrared diode laser sensors for the steelmaking industry. Dave's retirement and Sarah's new job as Combustion Chemistry Department Manager culminated 8 years of fruitful collaboration developing diode lasers for use in high-temperature industrial environments.



Professor Franz Winter (far right) of the Vienna University of Technology, Austria, visited (l-r) Linda Blevins, Chris Shaddix, and Jeff Murphy between August 20 and October 12. The group collaborated to examine nitrogen chemistry during pulverized coal char oxidation. Experiments were performed using the Sandia Multi-Fuel Combustor.

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Velocity Measurements Show Unexpected Source of Turbulence in High-Speed, Direct-Injection Diesel Engines

High-speed, direct-injection (HSDI) diesel engines have been identified as one of the most promising technologies for improving fuel economy and reducing CO₂ emissions of passenger vehicles. However, current engines will have to be significantly improved to meet the required NO_x and particulate emissions levels. Such improvement will require significant advances in the in-cylinder combustion process. One strategy for reducing emissions is to maintain high-levels of in-cylinder turbulence because high turbulence, with its associated high mixing rates, promotes the ‘burn-out’ of particulates (and remaining unburned fuel) before the dropping temperature associated with the expansion process begins to limit the combustion.

Paul Miles and Marcus Megerle, a graduate student from the University of Michigan, in conjunction with researchers at the University of Wisconsin, have shown that significant turbulence enhancement is present early in the expansion stroke and is predominantly due to the formation of an unstable momentum distribution. This source of turbulence has not previously been identified in engines. Their work combined experiments with modeling to clarify the effects of flow swirl on the in-cylinder turbulence. Increased flow swirl is one method by which engine designers attempt to maintain high turbulence. Until now, the specific mechanism for how flow swirl might enhance turbulence, other than by generally increasing velocity gradients, has not been identified. Indeed, some researchers have even argued that swirl provides an overall stabilizing influence, thereby reducing flow turbulence.

Paul and Marcus measured radial and tangential velocity under three operating conditions: motored, ‘injected’,

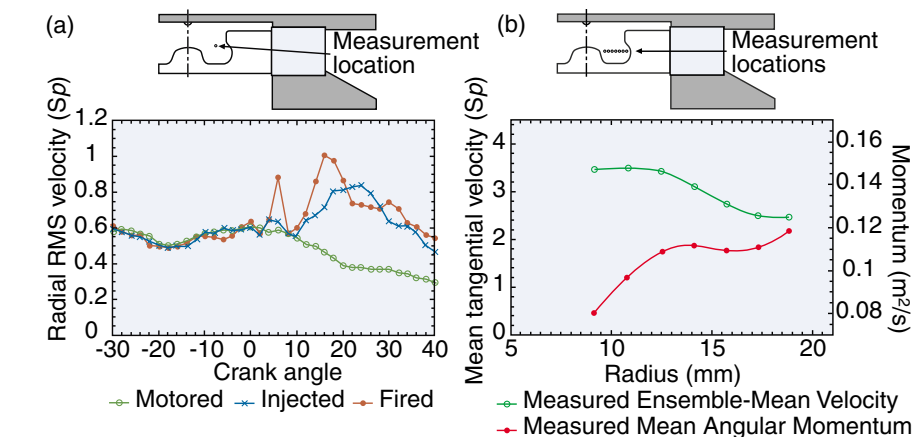


Figure 1. a) RMS fluctuating velocity normalized by the mean piston speed Sp measured for motored, injected, and fired engine operation. Under fired operation, the turbulence spikes early (6 CAD), returns to a level typical of motored operation, and then peaks at 18 CAD, an increase that is unaccounted for in current engine models. Under injected operation turbulence also increases confirming that significant turbulence is generated even in the absence of combustion. b) Radial profiles of the mean tangential velocity and angular momentum, demonstrate the formation of an unstable momentum distribution.

and fired. For injected operation, fuel is injected but no combustion occurs. The measured velocity fluctuations are shown in Figure 1. For fired operation, a brief increase in turbulence is seen during the peak heat release (about 6 crank angle degrees or CAD), but velocity fluctuations quickly drop back to levels typical of motored operation (no fuel injection or combustion). Later, turbulence again increases to a peak at 18 CAD that is nearly double the motored level. Current engine turbulence modeling does not capture this second, significant increase.

The most likely contributors to the increased turbulence were buoyant production, associated with density variations in the strong, swirl-induced radial pressure gradient, or an unstable angular momentum distribution within the cylinder. To generate an unstable momentum distribution, the tangential velocity must decrease rapidly with increasing radius, such that a negative radial gradient in angular momentum exists. Although this may seem implausible, a radial profile of tangential velocity—with a negative angular

momentum gradient—was measured at 21 CAD, shortly after the second peak in turbulence seen in Figure 1.

Modeling studies have helped clarify the mechanisms responsible for the formation of the unstable momentum distribution. Figure 2 depicts the mean velocity field, computed at 18 CAD, for motored, injected, and fired engine operation. The motored velocity field demonstrates that although the r - z plane flow structures are able to convect the high tangential velocity fluid to the bottom of the bowl, they are not sufficiently strong to transport this fluid inward and form an unstable angular momentum distribution. The fuel injection process, however, entrains low angular momentum fluid from the center of the bowl into the fuel jets, and deposits it at the bowl periphery. The computed results predict that this process of ‘pumping’ angular momentum forms an unstable momentum distribution even when combustion is inhibited. Turbulence measurements under injected operation, also shown in Figure 1, confirm that significant turbulence is

generated even in the absence of combustion. Buoyant production, therefore, plays a relatively minor role in generating the enhanced turbulence levels.

The significant turbulence enhancement observed in the early portion of the expansion stroke is thus shown to be predominantly due to modification of the in-cylinder flow by the fuel-injection process, through the formation of an unstable momentum distribution. Further clarifying this turbulence generation mechanism, and understanding how it varies as flow swirl and other engine parameters are varied, is a necessary step towards the intelligent optimization of HSDI diesel engines.

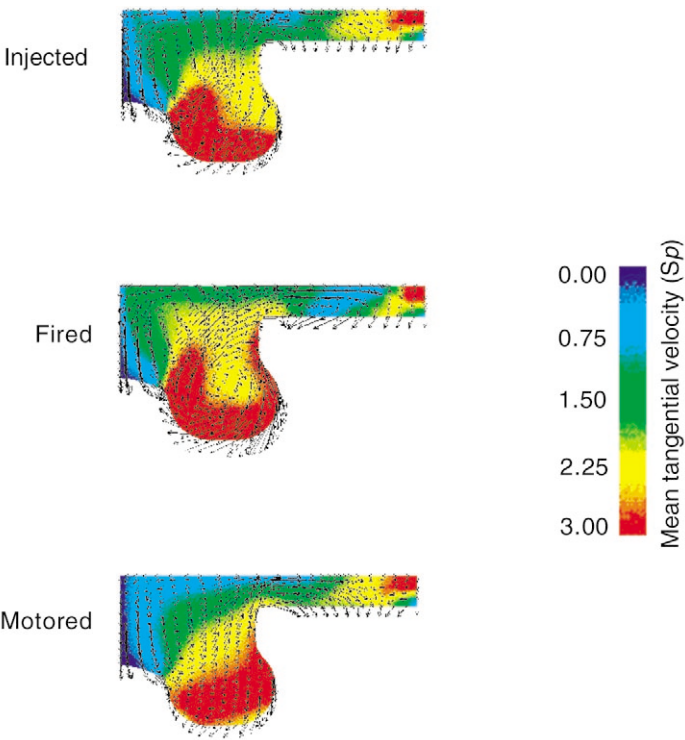


Figure 2. Full-field depiction of the three velocity components, as computed by collaborators at the University of Wisconsin. The vectors represent the velocity in the r-z plane, while the false-color background indicates the magnitude of the tangential velocity.

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